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The cross-polarization of light by large non-spherical particles

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Abstract

The paper is devoted to a theoretical investigation of the cross-polarization ratio of scattered light for randomly oriented particles of various shapes. The cases of spheroids, cylinders and fractal particles are considered in detail. The particles are assumed to be much larger than the wavelength of the incident light. Calculations are performed using the Monte Carlo ray-tracing code. Absorption of light by the particles is neglected.

It was found that highly irregular fractal particles give the maximum ratio of the cross-polarization. Particles with a larger degree of symmetry (e.g. circular cylinders and spheroids) give smaller values. The highest values of the cross-polarization ratio lay in the backward hemisphere. They were smaller than 0.8 for all the cases considered.

1. Introduction

The characterization of particulate systems is of importance to many branches of modern science and technology [1–4]. The size of dispersed particles, their chemical composition, morphology, concentration and shape are all of interest [5]. They are studied using a great variety of modern experimental and theoretical techniques [2].

This paper is devoted to theoretical modelling of relationships between cross-polarization of scattered light and shape of scatterers. Experimental work in this direction [6, 7] has already shown the potential of the cross-polarization scattering for the characterization of non-spherical large particles.

Note that we define the cross-polarization scattering as scattering by any object or medium placed between crossed polarizers.

Calculations were performed using the Monte Carlo ray tracing code for non-absorbing particles which are much larger than the wavelength of the incident light [8, 9]. In the geometrical optics limit considered in this paper, the intensity of scattered light is proportional to the area of the particle in the plane perpendicular to the incident light beam [10]. However, we are concerned here with the ratio of intensities. Since both intensities increase with area, this ratio is insensitive to the size of the particles. This allowed us to concentrate on effects that are solely due to particle shape.

The real part of the refractive index was fixed equal to 1.5, which is close to values of various minerals and chemical substances [7].

2. The cross-polarization ratio and Mueller matrix

We will consider the cross-polarization of scattered light by a collection of randomly oriented particles placed between crossed polarizers. In particular, we will assume that the incident light is polarized perpendicular to the scattering plane that contains the incident and scattered beams. Also we suppose that the linear polarizer just before the receiver transmits only light polarized in the plane of scattering. The concentration of particles is assumed to be small, so that multiple light scattering and dense media effects can be neglected.

Now we have for the Stokes vector of light registered by a receiver:

$$\vec{S}_{\mathrm{VH}} = \hat{M}_{\mathrm{H}} \hat{F} \hat{M}_{\mathrm{V}} \vec{S}_0 \qquad (1)$$

where $\vec{S}_0$ is the Stokes vector of the incident unpolarized light, $\hat{M}_{\mathrm{V}}$ is the Mueller matrix for the polarizer, placed before

the scattering medium, and $\hat{M}_H$ is the Mueller matrix for the polarizer placed after the medium. $\hat{F}$ is the Mueller matrix of randomly oriented isotropic non-spherical particles, which has the following general form [11]:

$$\hat{F} = \begin{pmatrix} F_{11} & F_{12} & 0 & 0 \\ F_{12} & F_{22} & 0 & 0 \\ 0 & 0 & F_{33} & F_{34} \\ 0 & 0 & -F_{34} & F_{44} \end{pmatrix} \quad (2)$$

Note it follows for spherical particles [11]: $F_{11} = F_{22}$ and $F_{33} = F_{44}$. The vertical and horizontal polarizers are characterized by the following matrices, respectively [12]:

$$\hat{M}_V = \frac{1}{2}\begin{pmatrix} 1 & -1 & 0 & 0 \\ -1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \quad (3)$$

and

$$\hat{M}_H = \frac{1}{2}\begin{pmatrix} 1 & 1 & 0 & 0 \\ 1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \quad (4)$$

Taking the product of matrices (2)–(4) and the vector $\vec{S}_0$ as in equation (1), we obtain for the first component of the Stokes vector $\vec{S}_{VH}$:

$$I_{VH} = \tfrac{1}{4}(F_{11} - F_{22})I_0, \quad (5)$$

which is the scattered light intensity. Here, I_0 is the intensity of incident light beam, which is the first component of the vector $\vec{S}_0$.

Let us consider now the same medium but placed between vertical polarizers. Then, we have instead of equation (1),

$$\vec{S}_{VV} = \hat{M}_V \hat{F} \hat{M}_V \vec{S}_0 \quad (6)$$

and, correspondingly,

$$I_{VV} = \tfrac{1}{4}(F_{11} - 2F_{12} + F_{22})I_0 \quad (7)$$

instead of equation (5).

Now we take the ratio of the intensities given by equations (5) and (7):

$$R = \frac{F_{11} - F_{22}}{F_{11} - 2F_{12} + F_{22}}, \quad (8)$$

which is called the cross-polarization ratio. This parameter can be easily measured experimentally [6, 7]. Theoretical approaches, on the other hand, are concentrated mostly on calculations of the matrix $\hat{F}$, which can be applied to a more broad range of experimental situations [4, 8, 9]. We will study the dependence R on the shape of the particles, having in mind possible applications to particle shape retrieval from optical measurements.

First of all we note that for spheres $F_{11} = F_{22}$ and $R = 0$. Thus, non-zero values of R mean the presence of non-spherical particles in the system. We would, however, like to go further and consider the possibility to distinguish disperse media with particles of different shapes by measuring curves $R(\theta)$, where θ is the scattering angle. We will consider cylindrical particles (with circular and hexagonal bases) and prolate spheroids randomly oriented in three-dimensional space. Mueller matrices for such particles can be found using exact electromagnetic theory [1, 4, 10], but computations become unstable for large scatterers with average size $a \gg \lambda$, where λ is the wavelength of the incident light. The characterization of such large particles is, however, of importance for a number of technological applications [2, 6, 7]. Therefore, to solve the problem we employ the ray-tracing approach, which is valid as $\lambda/a \to 0$. The code we use is described in detail by Macke [8]. Also, we assume that the particles are non-absorbing.

It is known [10] that the intensity of scattered light for a large ($\lambda/a \to 0$) non-absorbing particle is proportional to the area of the particle in the plane, perpendicular to the incident beam propagation direction in the geometrical optics limit. In reality, there are deviations [10] from such a behaviour for selected angular regions (e.g. forward, backward, and rainbow scattering). We neglect these deviations here, however.

We are concerned with ratios of intensities in this paper. Since these intensities both increase approximately with area, the ratio is insensitive to size. Experimental evidence for this is given by Card and Jones (1999). They found that for crystals of various types with sizes between 6.8 and 381 μm, the cross-polarization ratio has a good correlation to roundness with only a narrow spread. There was little indication of size sensitivity. This fact allows us to average over orientation distributions only and avoid averaging over the particle size distributions.

Note that many substances only weakly absorb light in visible [7]. Also it follows for the forward scattering zone: $R \to 0$ as $\theta \to 0$ independently of the shape and size of particles.

3. The results of calculations

The dependence $R(\theta)$, found from ray-tracing calculations, for large particles ($a \gg \lambda$) of different shapes is presented in figures 1(*a*)–(*c*). We assumed that the refractive index of the particles $n = 1.5$, which is close to the values used in the experiments [7]. Numbers on the figures show the aspect parameter, which is equal to the ratio of the length of cylinder to its diameter or the longest axis of a spheroid to the smallest one. The diameter of a hexagonal cylinder corresponds to the diameter of a circle around its hexagonal base. We see that the behaviour of the curves is quite different for particles of different shapes.

The largest value of R is reached in the region of scattering angle $\theta_{max} \approx 170^\circ$ for circular cylinders (see figure 1(*a*)). θ_{max} depends only weakly on the aspect parameter. The value of R is at maximum for smaller angles in the case of hexagonal cylinders (see figure 1(*b*)), where $\theta_{max} \approx 160^\circ$ (except the case of the aspect parameter $\xi = 1$, where $\theta_{max} \approx 170^\circ$ as for circular cylinders). It follows from figure 1(*a*) that the value of R for more elongated circular cylinders is generally larger than for more compact particles at $\theta < 140^\circ$. For angles $\theta > \theta_{max} \approx 170^\circ$ the opposite is true.

Hexagonal cylinders (see figure 1(*b*)) behave generally in a similar way to circular cylinders, except at angles $\theta < 60^\circ$, where R for circular cylinders is small (typically, smaller than 0.05). Note that the minimum at $\theta \approx 40^\circ$ for hexagonal

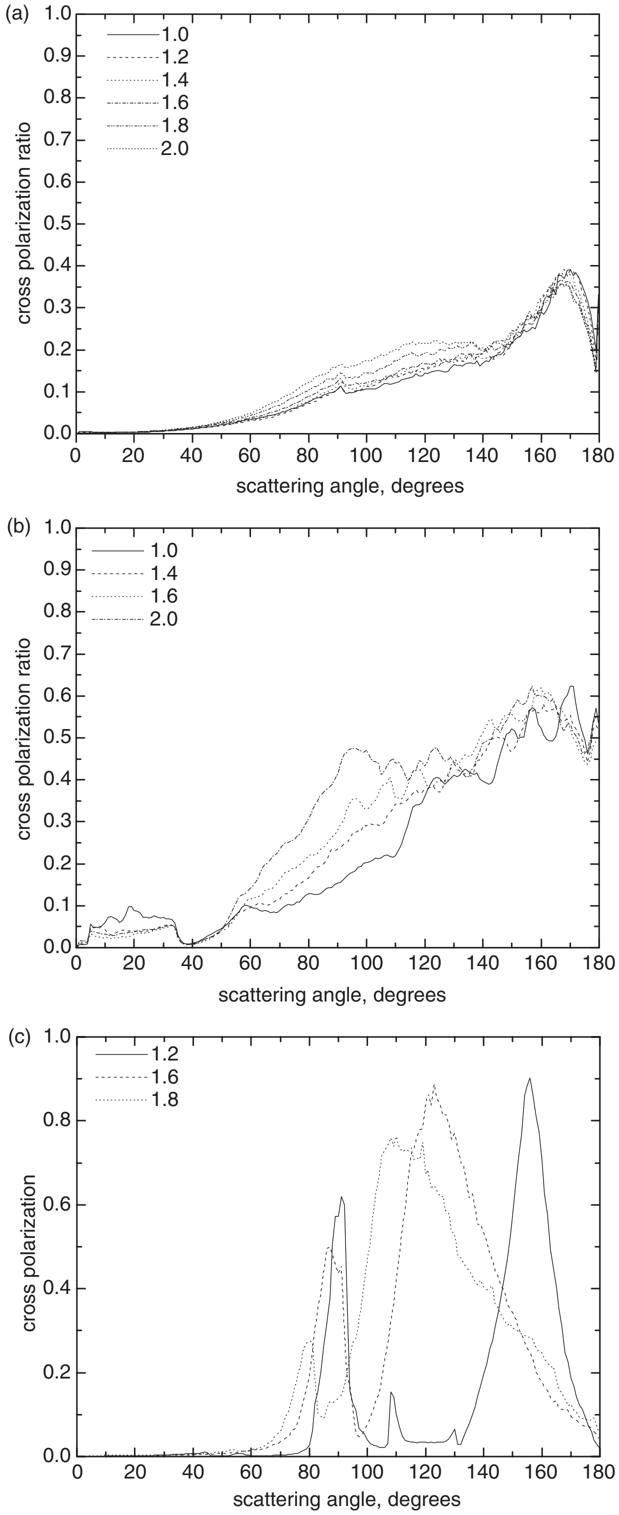

Figure 1. The dependence of $R(\theta)$ for randomly oriented circular cylinders (*a*), hexagonal cylinders (*b*) and prolate spheroids (*c*) at $n = 1.5$. Numbers on figures show the corresponding aspect ratios.

cylinders is related to a halo phenomenon, which is absent for circular cylinders. Generally, the behaviour $R(\theta)$ for circular cylinders is smoother than for hexagonal ones, where several maxima can be identified. Also generally the cross-polarization ratio is larger for hexagonal cylinders than for circular. This is due to their lower symmetry.

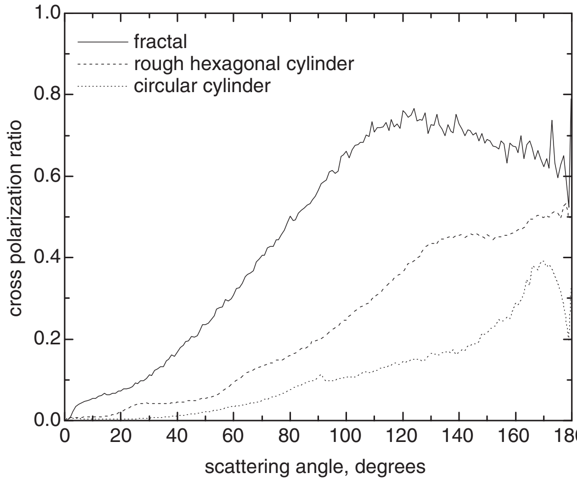

Figure 2. The dependence $R(\theta)$ for randomized Koch fractals, rough hexagonal cylinders and circular cylinders at $\xi = 1$ and $n = 1.5$.

The behaviour for spheroids is quite different to that of cylinders (see figure 1(*c*)). In particular, $R \approx 0$ at $\theta < 60^\circ$. The function $R(\theta)$ for ellipsoids is characterized by two strong peaks at each value of ξ. The distance between the peaks decreases with ξ, and so do their intensities. The peaks are positioned at smaller scattering angles for larger ξ. Outside the peaks the cross-polarization ratio is quite small.

These peaks are related to rainbow scattering by spheroids [3]. Clearly, they are washed out if one considers the averaging of results with respect to the aspect ratio. This will produce a curve similar to that presented in figures 1(*a*) and (*b*) with the maximum at the backward hemisphere.

We contrast the curves for circular and hexagonal cylinders with the same value of the aspect ratio $\xi = 1$ in figure 2. The ratios R for hexagonal cylinders oscillate with θ (see figure 1(*b*)). To diminish these oscillations we have used the model of rough hexagonal particles in figure 2. The roughness was modelled by averaging the calculated results with respect to the normal to the crystal surface, which was tilted randomly around its original direction, as described by Macke *et al* [9]. The zenith tilt angle was chosen randomly with equal distribution between zero and Ξ degrees. The value of Ξ was taken to be equal to 27°. We found that larger angles almost do not change the result of averaging. The azimuth tilt angle was randomly chosen from the full range of possible angles $[0, 2\pi]$. Comparing figures 1(*b*) and 2, we see that roughness leads to the disappearance of the halo around 40°. Also the curves become more smooth and any secondary maxima disappear. The function $R(\theta)$ generally increases with θ, not producing a sharply defined maximum. It follows that the value of R for hexagonal rough particles is larger than those for circular cylinders at all scattering angles.

The curve $R(\theta)$ for a randomized Koch fractal particle (see [9] for definitions) is also given in figure 2. The small-scale oscillations on this curve are due to the statistical noise of the Monte Carlo code. Note that light scattering by such particles represents quite well the main features of the scattering phenomenon for systems of irregularly shaped particles [3, 8, 9]. We see that randomized Koch fractals give the largest values of R among all the particles considered here. This suggests that irregularly shaped particles (even

when compact) give larger values of R as compared to particles of regular shape. Note also a broad maximum at the scattering angle close to 120° for fractals, where R is almost constant. Such behaviour of $R(\theta)$ around $\theta \approx 120°$ has been found experimentally for a large variety of irregularly shaped particles [7].

4. Conclusions

We have studied the cross-polarization of light by particles of different shapes (both regular and irregular) using the ray tracing approach described elsewhere [8]. It was found that irregular particles produce the largest values of the cross-polarization ratio as compared to particles with regular shapes. The function $R(\theta)$ has a maximum in the backward hemisphere. Positions of this maximum θ_{max} (and also $R(\theta_{max})$) can be used as indicators of particle's shape.

The values of the cross-polarization ratio for the particles studied are smaller than 0.8 at all scattering angles. The only exception is spheroids, where $R(\theta)$ may be larger than 0.8 around rainbow peaks. However, as was mentioned before, sharp spikes for ellipsoids are reduced due to the possible dispersion on their aspect parameter for natural media. Also, the value of $R(\theta)$ at spikes could be incorrect due to the inability of geometrical optics to describe rainbow phenomena [10]. So it is interesting to see in the experiment whether the value of $R > 0.8$ can be reached by any large non-absorbing particles either with regular or irregular shapes.

Also note that for fractal particles, the cross-polarization ratio is almost constant in the angular range 115–130°. Such behaviour was reported earlier by Card and Jones for the case of irregularly shaped particles [7]. Thus, we see that a fractal particle model captures some important features of the cross-polarization ratio curves for irregularly shaped particles.

The calculations presented here can serve as a basis for an interpretation of the curves $R(\theta)$, obtained from polarization measurements for various disperse media with large scatterers.

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